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DEMONSTRATION OF THE UREX SOLVENT EXTRACTION PROCESS WITH DRESDEN REACTOR FUEL SOLUTION

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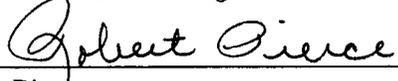
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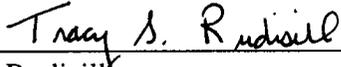
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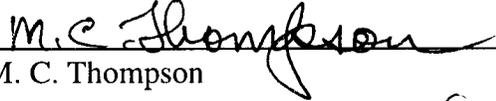
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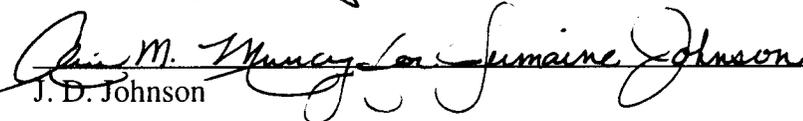

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Abstract

A solvent extraction process to recover uranium and technetium from solutions of irradiated commercial reactor fuel while sending the plutonium to waste with the fission products and higher actinides was tested with actual fuel solution. Demonstration of the uranium extraction (UREX) process at baseline conditions showed that the process meets all goals for recovery and decontamination. The goals for the UREX process are to produce a U product that is Class C low level waste or lower, recover > 99.9 % of the U and >95 % of the Tc, reject >99.9 % of the Pu to the raffinate, and maintain the U concentration in the Tc product <0.01 % of the Tc. All three tests showed Cs being lower than the Class A limit of 1 Ci per cubic meter. Strontium was low Class B for the first two tests and very close Class A in the third test (the Sr was below the detectability limit of the method in the third test). In fact, the third test resulted in a TRU isotope concentration of only 16 nCi per gram of U. The U losses to the Tc and raffinate streams totaled 0.011 % of the U fed to the process in the first test. U loss to the raffinate was 0.016 % in the second test.

Tc losses to the U stream were 1.2 % in the first test and 0.1 % in the second test while losses to the raffinate were so low they could not be measured in the first test. Data is not available yet for the other two tests. These results indicate that >95 % of Tc can be recovered from the feed.

Loss of Pu and other actinides to the Tc and U product streams was <0.02 % in all three tests of the baseline flowsheet with > 99.98 % going to the raffinate.

Introduction

Transmutation of waste is being developed to address disposal of commercial nuclear fuel and improve the performance of the geologic repository¹. The transmutation program will separate commercial fuel into (1) a transuranium (TRU) product stream that will be further processed, converted to fuel, and transmuted by fissioning to generate electrical power, (2) separate technetium-99 (⁹⁹Tc) and iodine-129 (¹²⁹I) streams, which will be converted into targets for transmutation to short-lived nuclides, and (3) a uranium product (UP) stream that meets the criteria for Class C low-level waste.

The Plutonium and Uranium Extraction (PUREX) process is a mature solvent extraction process for irradiated nuclear fuel that was designed to recover plutonium (Pu) and U. A variation of the PUREX process was conceived to provide the ability to treat the large quantities of irradiated spent fuel and to provide the selectivity required for the process. The PUREX process was changed so that only U and Tc are extracted and the TRU isotopes go to the aqueous raffinate along with the fission products. This Uranium Extraction process is called UREX (see Figure 1). The U solution from UREX is converted to uranium trioxide and the raffinate is evaporated and calcined to produce an oxide product that can be further treated by pyrochemical processing to separate the TRU isotopes from fission products. The raffinate could also be fed to other aqueous processes that recover Np and Pu together as well as cesium (Cs) and strontium (Sr). The Tc will be recovered from solution and converted into a target for irradiation. The process will use centrifugal contactors in order to minimize chemical and radiation damage to the solvent.

The ^{129}I will be volatilized during dissolution and recovered from the offgas.

The goals for the UREX process are to recover >99.9 % of the U and >95 % of the Tc in different product streams while rejecting >99.9 % of the TRU isotopes to the raffinate. The U product is to meet the requirements for Class C waste or lower for both fission products and TRU. Thus, the UO_3 product must contain <100 nCi of TRU per gram. The process must minimize the waste volume produced during processing. In order to minimize waste, all chemicals used in the process must be converted to gases during subsequent processing. In order to meet this requirement the process was designed to use acetohydroxamic acid (AHA) which complexes Pu(IV) and Np(IV) preventing them from extracting and reduces Np(VI) to inextractable Np(V)²⁻⁷. The flowsheet has been successfully tested with simulated solutions in batch and centrifugal contactor tests at Argonne National Laboratory (ANL).⁸ The tests reported here demonstrate the UREX process using solution from dissolved irradiated commercial reactor fuel from the Dresden Reactor.⁹

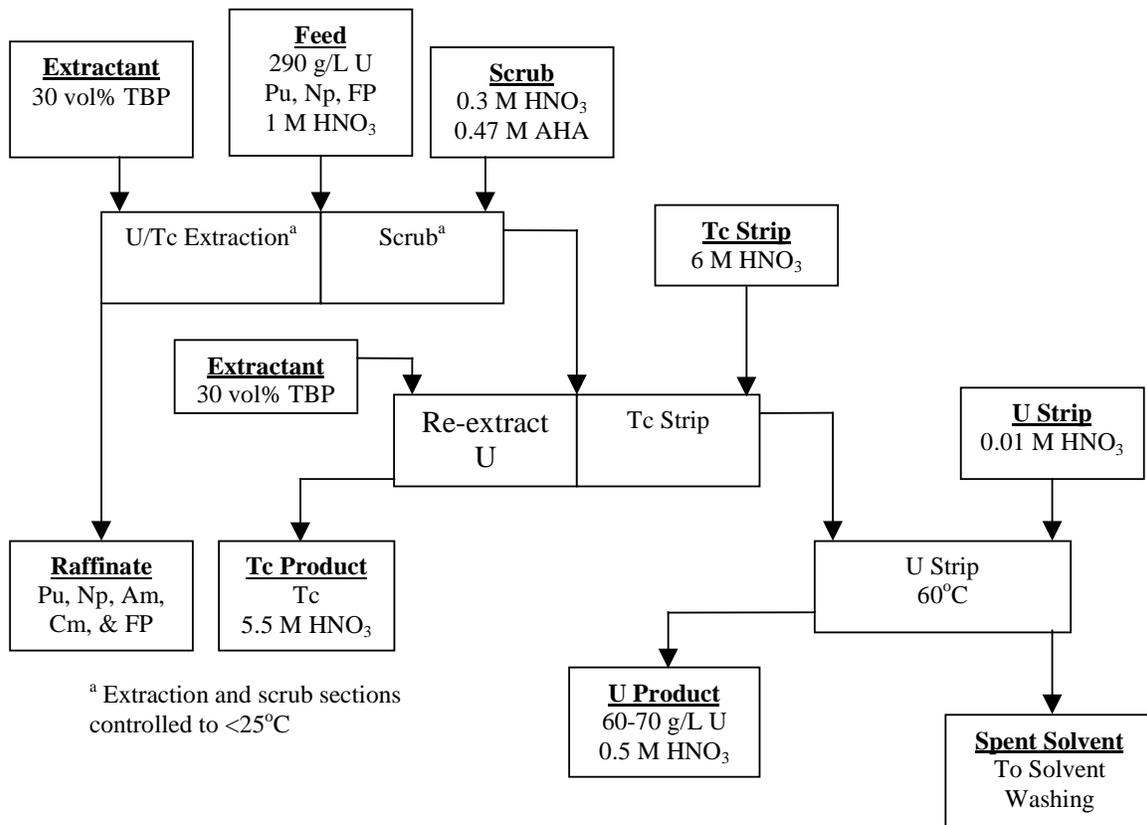


Figure 1: UREX Process Flowsheet

Experimental

Chemicals

Acetohydroxamic acid (AHA), 98 % pure, was obtained from Aldrich Chemical Co. and was dissolved in distilled water to produce a 0.94 M solution. Reagent grade HNO₃, 69-

71 wt% from Fisher Scientific Co. was diluted with distilled water to make acid solutions of 0.3, 0.6, 1.0 and 6 M for solvent extraction tests. The scrub solution for UREX tests was prepared by mixing equal volumes of 0.6 M HNO₃ and 0.94 M AHA together in the scrub feed tank immediately before starting to fill the contactors for the test. This was done to minimize acid hydrolysis of AHA.^{4,7}

The solvent, 30 vol% tributylphosphate (TBP), was prepared from 100 % TBP and n-paraffin obtained from Savannah River Site plant stocks. The n-paraffin is a mixture of C12 to C16 n-paraffins with low aromatic content. After preparation, the 30 vol% TBP solution was washed first with 5 wt% Na₂CO₃ (reagent grade chemical from Fisher Scientific Co.) to remove dibutylphosphoric acid and other impurities and then 0.01 M HNO₃ to re-acidify the solvent. The solvent was then stored in glass until needed for tests.

Solvent Extraction Feed

The solvent extraction feed solution was prepared by diluting solution obtained from the dissolution of Dresden Reactor fuel with 0.1 M and 3 M HNO₃.⁹ Table I shows the composition of the main components in the feed solution.

Table I: UREX Feed Solution Composition

Component	Analytical Results
Free Acid, M	0.84
U, g/L	302
Pu, g/L	1.96
Np, g/L	6.8 E-05
Am, g/L	1.18 E+09
Cm, g/L	
⁹⁹ Tc, g/L	1.68 E-04
¹³⁷ Cs, d/m/mL	2.16 E+10
⁹⁰ Sr, d/m/mL	2.26 E+10
¹⁵⁴ Eu, d/m/mL	1.99 E+08
¹⁵⁵ Eu, d/m/mL	3.99 E+07

There are uncertainties in the Pu concentration because dilution of the sample to remove from the shielded cell was so high that the uncertainty in the alpha counting was high. Additional analyses are incomplete at this time.

Solvent Extraction Equipment

The process equipment for extraction and Tc stripping consisted of a 32-stage, 2-cm annular centrifugal contactor apparatus installed in shielded cells at SRTC and used previously in demonstrations of a solvent extraction process for removal of cesium from SRS waste solutions.¹⁰ As a result of previous use, the contactor was flushed extensively and reconfigured for the UREX process. ANL personnel designed and fabricated the contactor stages. The number of stages in the shielded facility was only sufficient to

demonstrate U/Tc extraction and scrubbing and technetium stripping portions of the process. Four centrifugal contactor stages set up in a radiochemical hood will be used to strip the uranium from the loaded solvent. Figure 2 shows the process flow diagram for the arrangement in the shielded cells.

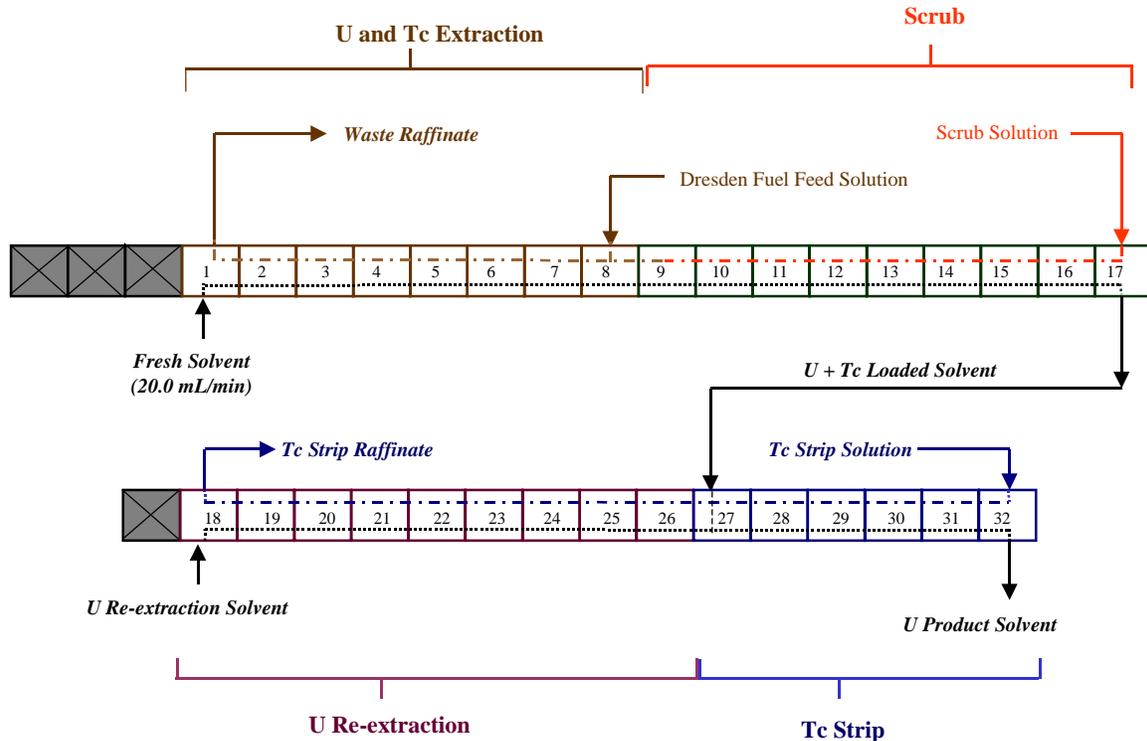


Figure 2: UREX Process Flow Diagram in Shielded Cell

Circulating cooling water maintained the temperature in stages 1-15 below 25°C to ensure Tc extraction. During the first two tests, a NesLab RTE-111 was used for temperature control. However, the NesLab chiller had problems maintaining temperature during the latter part of the second test. A new chilling system was provided by using a large chiller outside the cell to provide 6-8°C cooling water to a water bath in a dewar inside the cell. The dewar contained a heater to adjust the temperature of the water to 12-14°C. The cooled water was pumped through channels located on stages 1-15. Type K thermocouples were attached to the body of the contactor stages and connected to a computer for recording temperature data. The temperatures are measured on the surface of the contactor stages so the actual solution temperatures may be lower by 1-3 °C.

Positive displacement piston pumps manufactured by Fluid Metering, Inc. (FMI) were used for both organic streams and the scrub stream to the contactors. Masterflex peristaltic pumps from Cole Parmer Instrument Co. were used for the fuel feed and Tc strip streams. The initial set of Masterflex pumps contained microprocessors to control the flow rate. However, the Tc strip pump failed after use in the first two tests and was replaced with a pump with analog control. All pumps were calibrated prior to introduction into the shielded cell. All pumps except the scrub feed pump were operated

under manual control throughout the test. The scrub pump rate was controlled from the computer using the flow instrumentation from previous testing, but the flow rate was offset due to the difference in scrub stream properties. Figure 3 shows the layout of equipment in the shielded cell.

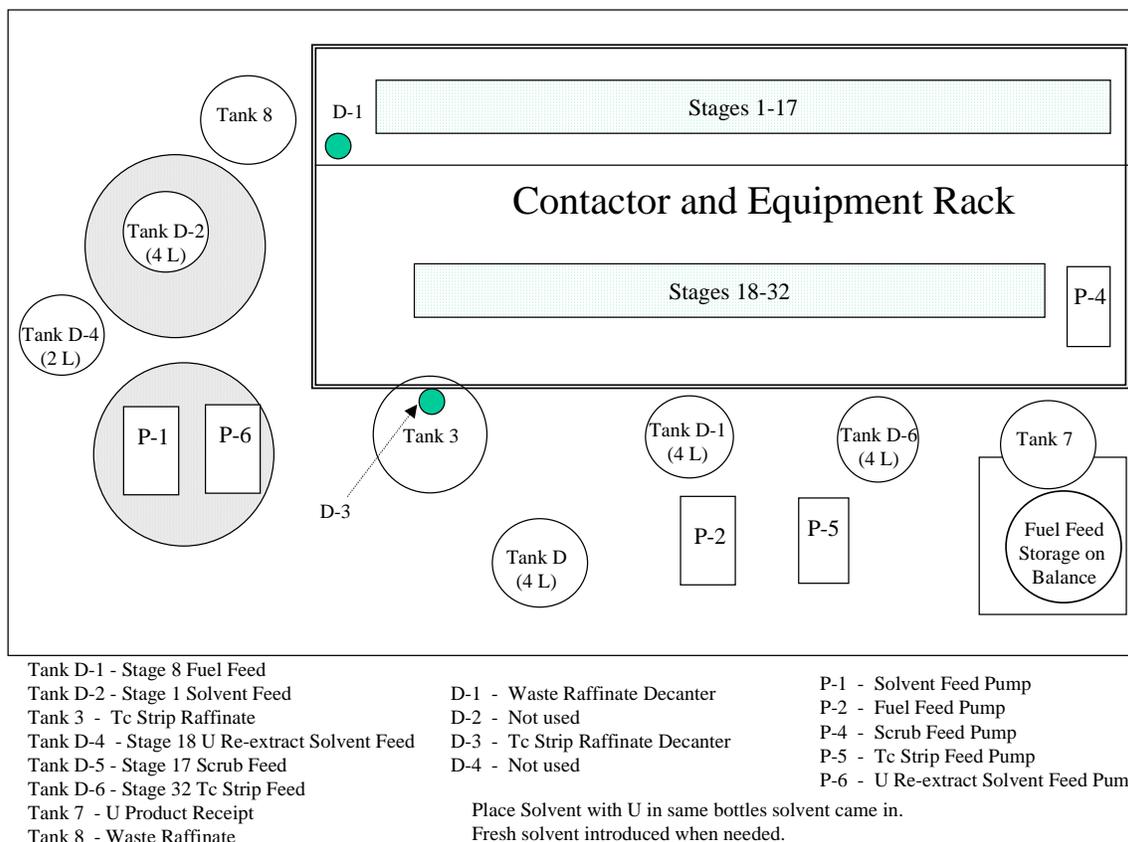


Figure 3: UREX Equipment Arrangement

Feed tanks were 2 or 4-L capacity graduated cylinders with a burette attached to the side that can be isolated from the main tank by a stopcock when checking flow rates (see Figure 4). The inlet to the pump was attached to the standpipe that is shown behind the burette. Manual adjustment of the feed pump speeds occurred as necessary based on the flow measurements. In general, the solvent flow rate held constant once set at the desired flow rate. Most differences can be attributed to the difficulty in observing the meniscus in the side arm due to pulsation from the FMI pumps. The uncertainty of the readings was estimated to be +/- 5 %. The uncertainty of the readings for the feed and Tc strip was lower because the meniscus did not pulsate as much.

To initiate an experiment, the rotors were started and then flow of the aqueous streams, surrogate feed (1 M HNO₃), scrub (0.47 M AHA in 0.3 M HNO₃), and strip (6 M HNO₃), was initiated. Aqueous flows were continued until flow was observed exiting into the raffinate and Tc product decanters. At this point, the solvent and U re-extract pumps were started and operated until organic was observed exiting stage 32. The feed tank was then emptied of 1 M HNO₃ and the feed solution pumped into the tank with a transfer pump.



Figure 4: Feed Tank for UREX Solvent Extraction Tests

During tests, the following items were monitored. The frequency at which these items were monitored is shown in parentheses.

- Motor rotation (15 minutes)
- Liquid in standpipes (15 minutes)
- Feed and collection tank levels (30 minutes).
- Decanter levels (1 hour)
- Temperature trends (1 hour)
- Measured process stream flow rates (1 hour or as needed).

Samples were collected by placing sample bottles under the outlet points of the continuously flowing streams. Samples were taken hourly during the simulant and first two tests and every two hours during the third test.

At the end of each test, researchers stopped the motor rotation and feed pumps simultaneously to minimize disruption of the contents of each stage for the post-test stage samples. Drain valves on each contactor stage allowed removal of each stage's contents at the end of the test.

Solvent Extraction Tests

Hydraulic Testing

Hydraulic tests were performed with the contactor system to determine that the desired flow rates would result in stable operation. Both the feed and Tc strip solutions were 1 M HNO₃ instead of the flow sheet solutions of 290 g/L U in 1 M HNO₃ and 6 M HNO₃, respectively. This was done to reduce the volume of waste produced and should be conservative relative to hydraulic operation due to the lower density of both solutions compared to the actual feed solutions. The scrub solution was 0.3 M HNO₃. The planned conditions for the tests were:

- Feed – 5.7 mL/min
- Solvent – 20 mL/min
- Scrub – 3.3 mL/min
- Tc Strip – 20.0 mL/min
- Tc Re-extract – 5.5 mL/min

However, a short time after start of Tc re-extract flow, the decanter filled with solvent indicating the strip contactor stages would not operate at the combined flow rate of 45.5 mL/min. Failure to operate at this total flow rate was unexpected since ANL had used similar flows during a simulate test done earlier. Operation of the rotors was examined and stage 21 was found to be suspect. The stage 21 rotor was subsequently replaced. The contactors still did not operate correctly so all flow rates were lowered by 25 % to 4.28, 15.0, 2.48, 15.00, and 4.13 mL/min, respectively. The contactor hydraulics worked well so the flows were increased to 4.85, 17.0, 2.81, 17.0, and 4.68 mL/min, respectively. The contactors were operated again for several hours and then the U re-extract flow was increased from 4.68 to 5.53 mL/min. No hydraulic problems were observed at these flow rates and no further hydraulic testing was done.

Simulant Test

A test of the UREX flowsheet was done with simulated feed solution prior to the first test with Dresden fuel solution. Two liters of simulant solution were prepared by dissolving 684 g of U₃O₈ (depleted U) in 1.5 L of 5 M HNO₃ containing 1.2 g of sodium perrhenate (NaReO₄, as a non-radioactive surrogate for Tc). Water was added to increase the volume to 1875 mL prior to adding 108 mL of 40 g/L Pu nitrate in 1 M HNO₃. The solution was then diluted to 2 L with water. The Pu was weapons grade material, which has a different isotopic composition than the Dresden fuel Pu. Analytical results on the feed solution showed 287 g/L U, 1.03 M free acid, 0.4 g/L Re, and 2.13 g/L Pu.

Dresden Fuel Solution Tests

Three tests of the UREX process were made with the Dresden fuel feed. The flow rates and concentrations of AHA and acid were determined from calculations with the AMUSE computer program by ANL personnel. In each test, the flow rates were measured at the start of feed flow, 45 minutes after starting and every 60 minutes thereafter. Samples of all 3 end streams were taken every hour after the first 2 hours of operation. During the first 2 hours of operation, the waste raffinate was sampled every 30 minutes, the Tc product and U-loaded solvent streams were sampled at 1, 1.5, and 2

hours. The organic U product samples were stripped with sodium carbonate and re-acidified prior to submitting for analysis. The treatment for stripping and re-acidification resulted in a dilution by a factor of 5 for samples submitted for analysis except for U where the dilution was by a factor of 10. Waste raffinate samples were diluted (1000-5000 times) as needed to allow removal from the shielded cell for analyses. Samples from stages 1, 3, 5, 7, 9, 11, 13, 15, 17, 18, 20, 22, 24, 26, 28, 30 and 32 were collected for later separation. The stage samples were placed in separatory funnels to allow the phases to completely separate prior to sampling for subsequent analysis.

Analytical Methods

All analyses were done by the Analytical Development Section at SRTC. Uranium product analyses were done spectrophotometrically on solutions adjusted to 1 M HNO₃ and 1-10 g/L U.¹¹ Lower concentrations of U were done either by inductively coupled plasma mass spectroscopy (ICP-MS) or laser fluorescence spectroscopy. Analyses of general elemental concentrations were done by inductively coupled plasma emission spectroscopy. Analyses for ⁹⁰Sr required separation prior to beta counting using liquid scintillation. Plutonium analyses were done by extraction with thenoyltrifluoroacetylacetone in hexane followed by alpha pulse height analysis. A second sample was spiked with Pu-239 and the same extraction and counting to determine the recovery efficiency. Technetium analyses were done by separation with ion exchange and beta counting.

Results and Discussion

UREX Solvent Extraction Testing

Simulant Test

Prior to using dissolved Dresden fuel solution, a test was made with solution simulating the expected acid, U, Pu, and Tc (using Re as a surrogate for Tc) concentrations in the Dresden fuel feed solution. The simulant test lasted for six hours. Table II shows the flowsheet and actual flow rates during the test. Note that the feed and scrub flow rates varied more than the other flow rates. The range for the solvent is probably the variability of the flow measurement. Note that flow rates of all aqueous streams appear to be slightly lower than the desired flows.

Figure 5 shows the flow rate ratios during the test. Note that the flow rate ratios are close to the desired ratios (the straight lines on the chart) except for the solvent to feed plus scrub ratio. That ratio was high because the scrub flow rate was low during much of the test. The low scrub flow is also shown in the scrub to feed ratio where the flow ratios are low until about 3.5 hours into the test.

Table II: Flow Rates for UREX Simulant Test

Stream	Flowsheet, mL/min	Actual Flow rate Range, mL/min
Extractant	17.0	16.8-17.0
Feed	4.85	4.65-4.79
Scrub	2.81	2.50-2.76
Tc Strip	17.0	16.75-16.85
U Re-extract	5.50-6.00	5.92

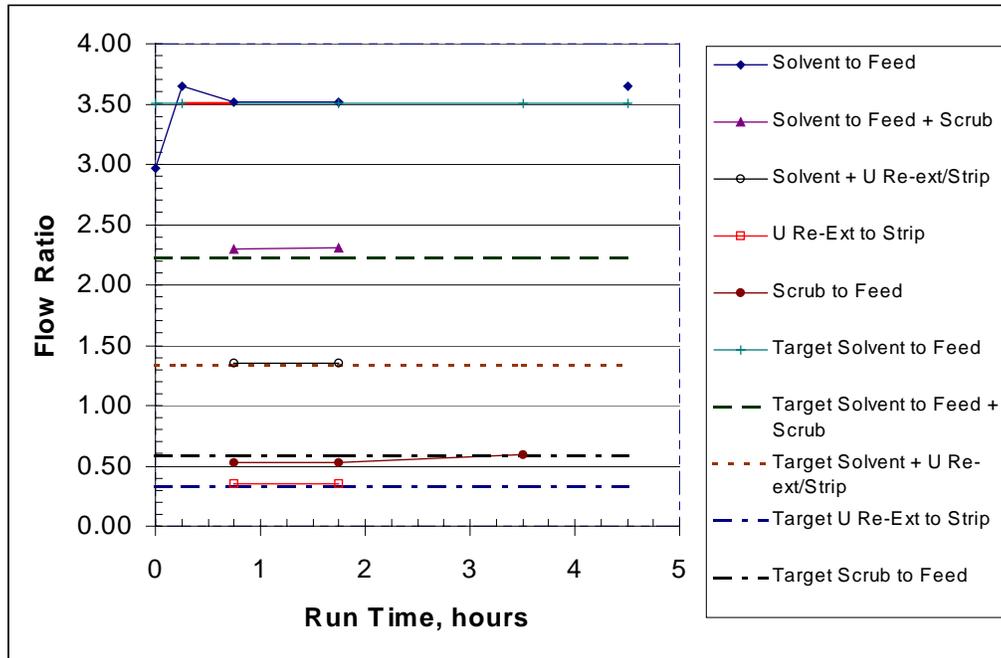


Figure 5: Simulant Test Flow Ratios

The test ran well hydraulically with <1% other phase contamination of any stream. Pu was rejected to the waste and U was recovered in the U product stream (see Table III). U was low in the Tc product and raffinate streams. Note that the U in the raffinate is well below 0.01 % of that in the feed. However, the U in the Tc product stream would be higher than the 0.01 % of the Tc concentration. The Pu decontamination factor for the U product is 137,000, which is high enough to produce a non-TRU product. However, Re chemistry is apparently different enough that the Re mostly went to the waste raffinate along with Pu; some went to the U product stream rather than stripping into the Tc product stream. Overall the simulant test showed the flowsheet could be operated hydraulically and the U product was not a TRU waste.

Table III: Mass Balance for UREX Simulant Test

Stream	Time of Operation, hrs	% U	% Pu	% Re
U Product	0			
	1	76.4	6.47E-04	2.34
	1.5	96.1	7.59E-04	2.92
	3	91.1	6.67E-05	2.46
	4	100.7	4.06E-04	2.83
	5	101.3	5.53E-03	2.98
	6	101.7	7.30E-04	2.61
Tc Product	0			
	1	No Data	1.00E-04	0.36
	1.5	No Data	7.08E-05	0.49
	3	No Data	6.19E-05	0.31
	4	No Data	6.39E-05	0.15
	5	1.22E-02	1.20E-04	0.09
	6	3.65E-02	4.03E-04	0.09
Waste Raffinate	0			
	1	No Data	37.5	25.56
	1.5	No Data	95.8	70.75
	3	No Data	114.5	93.51
	4	No Data	115.6	105.20
	5	9.67E-05	106.1	101.08
	6	9.13E-05	103.9	98.75

UREX Tests with Dresden Fuel Solution

UREX Hot Demonstration Test 1

Three tests were done with the 13 liters of solution obtained from dissolution of Dresden fuel. In the first test, inadequate mixing of the dissolver solution during dilution resulted in the feed solution being much higher in U (466 g/L) and acid (~1.4 M) than the flowsheet is designed to process. In addition, there was an accidental cessation of feed flow during the test and some other operational problems. Temperatures measured during the test varied as shown below.

Stages 1-15: 20 - 28 °C

Stages 16-17: 26 - 33 °C

Stages 18-32: 26 - 37 °C

The temperature in the extraction section should be 25 °C or less to ensure the extraction of Tc. Some of the scrub stages were above the desired temperature of 25 °C so it was possible that some Tc could be lost to the raffinate stream. The high scrub section temperatures were present in all the tests because the last scrub stages have no cooling other than the organic solvent containing the U. As a result of the problems encountered, analyses were not completed. However, some promising results were observed from the limited analyses. The test showed better control of the flow rate ratios than in the simulant test (see Figure 6).

Better control of flows and flow ratios gave relatively good decontamination of the U and Tc product solutions. The results of analyses during the test are shown in Table VI. Note that the U concentration in the U product should have been almost 90 g/L. However, little of the U appears to be in the raffinate. Almost none appears in the Tc product as expected with 6 M HNO₃ in the strip solution. There could have been some accumulation of U in the contactors. However, a calculation of the quantity of U that could be in the contactors is <50 grams (based on the volume per stage of 30 mL) relative to a 150-200 g U discrepancy in the mass balance. It is more likely that there is a dilution error during stripping of the U from the solvent. The U product meets low level waste criteria for Tc, Cs, Pu-241, and Sr, based on the data at the end of the test.

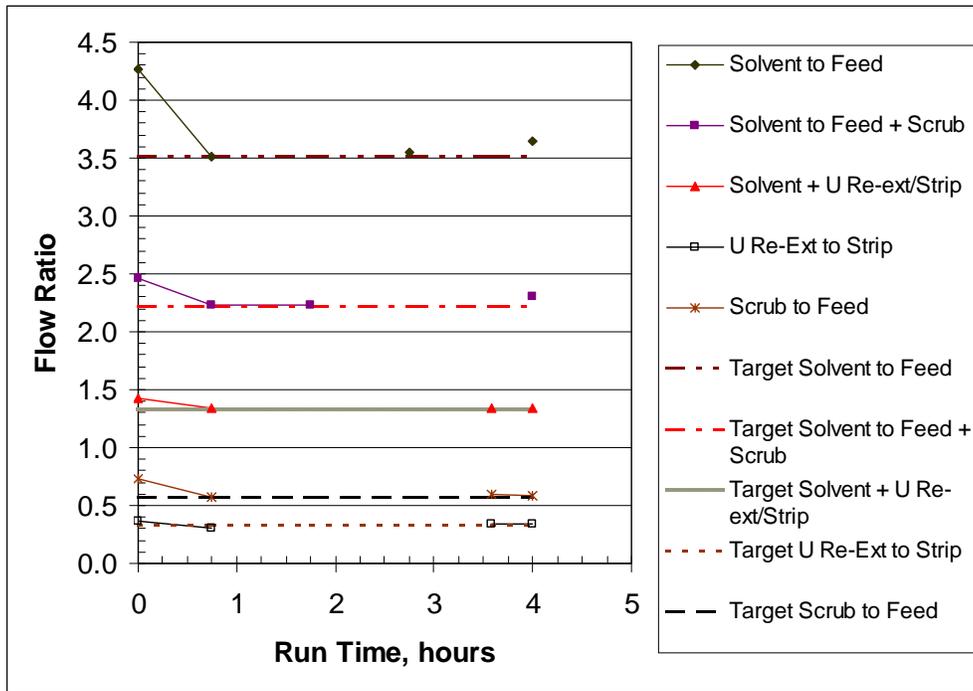


Figure 6: Flow Ratios for UREX Test 1

Table IV: End Stream Analyses from UREX Hot Demonstration Test 1

Stream	Time of Operation	U	Pu	Am	Cm	Tc	Cs	Sr
		g/L	d/m/mL	d/m/mL	d/m/mL	d/m/mL	d/m/mL	d/m/mL
U Product	0	13.1	2.60E+03	ND	ND	<2.1E+03	1.30E+03	8.10E+03
	1	36.2	1.89E+03	2.81E+03	189	<2.7E+03	3.30E+04	<5.2E+03
	1.5	33.7	2.19E+03	1.55E+03	179	<1.2E+04	2.34E+04	<5.2E+03
	2	35.5	1.34E+03	1.45E+03	292	<2.1E+03	1.72E+04	<5.2E+03
	6	31.9	1.14E+04	9.05E+03	465	<1.3E+03	1.09E+04	<5.2E+03
Tc Product	0	ND	5.32E+02	4.79E+02	165	2.20E+03	2.68E+05	3.57E+03
	6	0.0045	4.54E+02	4.57E+02	48	1.08E+06	6.90E+04	6.35E+03
Waste Raffinate	0	ND	7.94E+07	ND	ND	1.04E+04	1.39E+06	1.01E+06
	6	0.227	1.14E+09	1.02E+09	ND	3.07E+05	1.02E+10	1.52E+10

ND – No Data

However, the TRU isotope alpha is almost three times higher than the limit of 100 nCi/g U. If the U concentration in the product had been the expected 90 g/L, the TRU limit would be exceeded by only 10%. The Pu losses to the U and Tc product streams totals <0.01 % of the Pu in the feed. This result confirms that >99.9 % of the Pu was in the raffinate stream meeting one of the goals of the process. Total U losses to the Tc product and raffinate streams was 0.08 % of the U. Although this is higher than expected, it still meets the goal of >99.9 % recovery of U.

UREX Hot Demonstration Test 2

The second UREX test was run for 6 hours with stable hydraulic operation and few operational problems. Temperatures measured during the test varied as shown below.

Stages 1-15: 17 - 28 °C

Stages 16-17: 26 - 32 °C

Stages 18-32: 27 - 35 °C

During operation, personnel noted that the raffinate in the decanter was very dark in color as expected for the Pu AHA complex, which is a dark red. The color of the Fe AHA complex is a method for analysis of AHA concentration in solution.

Figure 7 shows the flow ratios over the time of the test. Note that the flow ratios did not match as well as in test 1. The solvent to feed ratio was high, which would lead to lower saturation of the solvent and possibly lower decontamination of product streams.

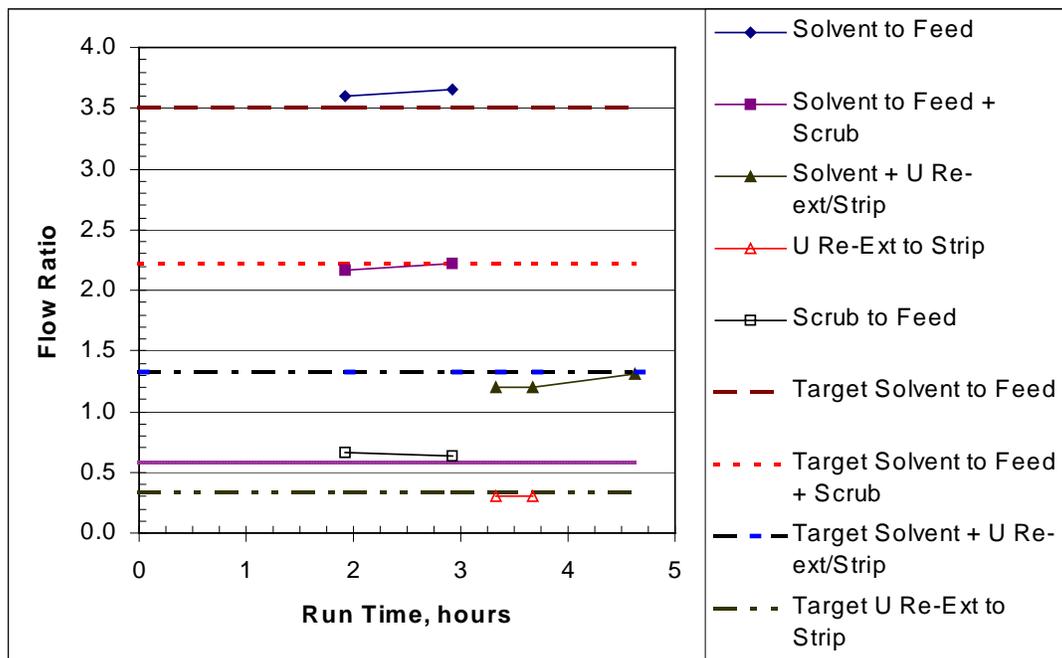


Figure 7: UREX Test 2 Flow Rate Ratios

Table V shows the analytical results for test 2 and Table VI shows the mass balances for the test. The acid concentration in the Tc product solution after 6 hours of operation was measured to be 5.21 M. The data show that U almost reached steady state concentration

in the U product after one hour of operation and was at steady state by 3 hours. That does not appear to be the case for the other nuclides. Pu and Am data seem to go up and down.

Table V : Analytical Results for Test 2

Stream	Time of Operation	U	Pu	Am	Cs	Tc	Sr
		g/L	d/m/mL	d/m/mL	d/m/mL	d/m/mL	d/m/mL
U Product	0	3.9	4.12E+04	1.16E+04	7.90E+04	2.11E+03	1.02E+04
	1	51.2	4.43E+03	2.85E+03	6.05E+03	2.72E+03	3.84E+03
	3	54.5	6.01E+03	0.00E+00	8.98E+03	1.15E+04	3.85E+03
	4	53.7	1.18E+04	1.53E+04	1.17E+04	2.07E+03	3.85E+03
	6	55.1	3.02E+03	5.07E+03	1.67E+04	1.29E+03	5.00E+03
Tc Product	0	<4.5E-03	NA	NA	1.53E+05	2.25E+04	2.10E+04
	2	<4.5E-03	2.41E+03	NA	1.28E+05	1.58E+06	1.46E+04
	3	<4.5E-03	2.31E+04	1.85E+04	4.48E+05	1.82E+06	2.30E+05
	4	<4.5E-03	8.41E+03	5.00E+03	3.34E+05	1.81E+06	1.02E+05
	6	7.9E-05	4.67E+03	7.53E+03	3.93E+05	1.45E+06	1.11E+05
Waste Raffinate	1	NA	6.72E+08	8.79E+08	1.77E+10	1.64E+06	NA
	2	NA	8.55E+08	6.57E+08	1.26E+10	5.84E+06	9.78E+09
	3	NA	1.04E+09	NA	1.93E+10	5.72E+06	1.30E+10
	4	0.0026	7.57E+08	NA	1.68E+10	1.45E+06	1.94E+10
	6	0.0297	7.97E+08	7.50E+08	1.89E+10	2.90E+06	1.47E+10

Table VI: Mass Balance Results for Test 2

Stream	Time of Operation	U	Pu	Am	Cs	Tc	Sr
		%	%	%	%	%	%
U Product	0	6	1.87E-02	5.13E-03	1.81E-03	<0.17	2.24E-04
	1	84	2.01E-03	1.26E-03	1.38E-04	<0.21	<.84E-05
	3	90	2.76E-03	NA	2.08E-04	<0.91	<8.5E-05
	4	89	5.42E-03	6.82E-03	2.71E-04	<.16	<8.5E-05
	6	91	1.38E-03	2.26E-03	3.86E-04	<.10	1.11E-04
Tc Product	0	<6.1E-03	NA	NA	2.86E-03	1.4	3.76E-04
	2	<6.1E-03	8.95E-04	NA	2.39E-03	101	2.43E-04
	3	<6.1E-03	8.68E-03	6.76E-03	8.48E-03	118	3.88E-03
	4	<6.1E-03	3.14E-03	1.82E-03	6.29E-03	116	1.71E-03
	6	9.8E-05	1.61E-03	2.52E-03	6.82E-03	86	1.71E-03
Waste Raffinate	1	NA	103	131	136	43	NA
	2	NA	131	98	97	154	72
	3	NA	157	NA	146	148	94
	4	0.0014	114	NA	127	38	141
	6	0.0161	120	110	143	75	107

NA - No analysis

It is unlikely to be contamination because the Cs, Tc and Sr data do not show the same effect. The U mass balance is lower than observed in the simulant test, but is within 10 %. The difference could be in measurement of the organic flow rates, which pulsed due to the piston pump action. Regardless of the low U mass balance, the U product from this test is lower than Class C waste for TRU and all fission products demonstrating that the UREX process meets that goal.

In addition, although the mass balance data indicate that less than 99.9 % of the U was recovered, less than 0.1 % of the U is in the raffinate and Tc product (0.016 %). Data on U in the Tc stream is very low at about 1 E-04 %.

The mass balance for the Tc product stream appears to go through a maximum and then decrease again. However, the average of the data from 2 through 6 hours is 105 %. The Tc behavior could also be a flow rate problem, but no flow measurements were made over the last four hours of the test. It should be noted that the Tc strip flow rate was high early in the test and efforts were made to reduce the strip flow closer to the desired flow rate of 17.0 mL/min. The raffinate also shows large amounts of Tc. The problem with the existing data is that the samples had very high very high dilutions (11,000 to 27,000) so minor contamination of the samples led to high Tc counts in the samples. The Tc product samples are much more reliable because no dilution was made on those samples prior to analysis. It appears from the average of the Tc product data that the goal of > 95 % Tc recovery was achieved in this test.

The data also show that 0.011 % of the TRU was in the two product streams demonstrating rejection of >99.9 % of the Pu to the raffinate. Therefore, this test demonstrated that all the goals for the UREX process were met.

UREX Hot Demonstration Test 3

The third test was done in two parts, 3A and 3B. Test 3A was 8 hours long with flow rates and feed stream flow rate ratios identical to the first two tests. Test 3B was run with different flow rates and flow ratios in both extraction and stripping.

Test 3A operated with stable hydraulics and no operational problems. Temperatures measured during the test varied as shown below.

Stages 1-15: 19 - 28 °C

Stages 16-17: 29.1 - 36.7 °C

Stages 18-32: 26.5 - 39.6 °C

The temperatures for stages 16- 32 were measured over the entire time that tests 3A and 3B were in progress. The temperatures for stages 1-15 were measured during the actual time of the test.

The flow rates of the feed streams were to be the same as for the previous two tests. Table VII shows the flow rates measured during the test 3A including the overall average and standard deviation. The feed flow rate was especially stable during the last portion of the test. Overall the standard deviation for three streams is less than 2 %. Such deviations are within the measurement error for flow rates. The other two streams had standard

deviations of 4% and 6%. The 6% deviation was for the scrub stream, which is greater deviation than can be accounted for by measurement uncertainty.

Table VII: Flow Rates Measured during Test 3A

Run Time, hours	Solvent Flow rate, mL/min	Feed Flow Rate, mL/min	Scrub Flow Rate, mL/min	Strip Flow Rate, mL/min	U Re-Extract Flow Rate, mL/min
0.58	16.80	4.61	2.87	17.17	5.86
0.67		4.67			
1.08		4.72			
1.75	16.80	4.79	2.95	17.07	5.83
2.75	16.60	4.83	3.36	16.96	5.23
3.75	16.90	4.82	3.04	17.07	5.95
4.92	16.40	4.85	2.81	17.07	5.83
6.17	16.80	4.82	2.95	16.75	5.88
7.00	17.10	4.82	2.85	17.28	5.88
7.95	17.30	4.85	2.99	17.07	5.87
Average	16.84	4.78	2.98	17.05	5.79
Std Dev	0.28	0.08	0.17	0.15	0.23

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Figure 8: Flow Ratios for Test 3A

Figure 8 shows the flow ratios over the time of the test. The solvent to feed ratio appears to vary around the target value. The solvent to feed plus scrub flow ratio is lower than the target due to the higher than desired scrub flow during much of the test. The higher scrub flow is also apparent in the scrub to feed ratio being higher than the target. The higher scrub flow rate gave higher decontamination for fission products and TRU than observed in the previous tests.

Table VIII shows the analytical results for test 3A and Table IX shows the mass balances for the test.

Table VIII: Analytical Data for Test 3A

Stream	Run Time	U	Pu	Am	Cm	Tc	Cs	Sr
		g/L	d/m/mL	d/m/mL	d/m/mL	d/m/mL	d/m/mL	d/m/mL
U Product	0	30.3	8.31E+03	7.93E+03	2.03E+03	NA	1.46E+04	1.67E+03
	2	58.9	6.68E+03	4.55E+02	1.17E+02	NA	2.31E+04	3.35E+03
	4	12.6	2.69E+03	4.30E+02	1.10E+02	NA	1.08E+04	6.70E+03
	6	62.7	8.60E+02	3.70E+02	9.49E+01	NA	2.34E+03	3.35E+03
	8	61.8	1.11E+03	1.09E+03	2.79E+02	NA	1.66E+03	3.35E+03
Tc Product	0	NA	1.50E+02	NA	NA	5.88E+05	4.56E+05	NA
	2	NA	1.20E+02	NA	NA	5.98E+05	1.03E+05	2.76E+03
	4	NA	4.50E+02	NA	NA	1.34E+06	1.28E+05	4.08E+03
	6	1.0E-05	2.11E+02	NA	NA	1.40E+06	7.42E+04	4.37E+03
	8	1.0E-05	8.54E+02	NA	NA	1.38E+06	5.72E+04	2.45E+03
Waste Raffinate	6	NA	1.09E+09	NA	NA	NA	NA	2.39E+10
	8	NA	1.63E+09	NA	NA	NA	NA	2.02E+10

NA - Not Available

Contamination of the U product from this test is the lowest of any of the tests and far lower than Class C waste limits for TRU and all fission products. The data demonstrate that the UREX process meets the goal for the U decontamination. The mass balance for U is quite good at 97.8 % in the last sample. The decontamination of the U product from fission products was so good that four liters of the organic was removed from the shielded cell for stripping U in a hood. The dose from that quantity of organic solvent was only 1 mrad/hr, indicating a very low level of gamma activity.

The mass balance for the Tc product stream appears to be poor with a maximum of about 70 % with the last two samples essentially identical. That may be indicative of analytical error in the feed analysis. A second sample was taken, but results are unavailable at this time. The data also show that 0.011 % of the TRU was in the two product streams demonstrating rejection of >99.9 % of the Pu to the raffinate. Thus, this test demonstrated that most of the goals for the UREX process can be met.

Table IX: Mass Balances for Test 3A

Stream	Run Time	U %	Pu %	Am %	Cm %	Tc %	Cs %	Sr %
U Product	2	92.1	1.81E-04	1.92E-04	5.75E-04	NA	5.00E-04	<7.0E-05
	4	19.8	1.71E-04	1.82E-04	5.45E-04	NA	2.36E-04	1.41E-04
	6	85.8	1.29E-04	1.37E-04	4.09E-04	NA	4.43E-05	<6.1E-05
	8	97.8	4.38E-04	4.65E-04	1.39E-03	NA	3.63E-05	<7.1E-05
Tc Product	2	NA	1.35E-04	NA	NA	34	2.09E-03	1.42E-04
	4	NA	3.57E-05	NA	NA	75	1.67E-03	6.34E-05
	6	1.2E-05	6.16E-05	NA	NA	77	1.18E-03	6.67E-05
	8	1.2E-05	2.53E-04	NA	NA	77	9.23E-04	6.24E-05
Waste Raffinate	6	NA	NA	NA	NA	NA	NA	170
	8	NA	118	NA	NA	NA	NA	144

NA - Not Available

Test 3B was a continuation of test 3A except that the flow rates were changed to provide a higher solvent to feed flow ratio and lower strip and U re-extraction flows to increase the concentration of U and Tc in their respective product streams. The flow rates were changed based on calculations done at CEA with the PAREX computer code by Dr. Pascal Baron and Dr. Binh Dinh.¹² New flow rates are shown below. Note that the solvent flow rate was the only flow that was held constant.

Table X: Comparison of Baseline Flow Rates and Flow Rates for Test 3B

Stream	Baseline Flows ML/min	Test 3B Flows mL/min
Feed	4.85	4.5
Solvent	17.0	17.0
Scrub	2.8	3.0
Tc Strip	17.0	8.5
U Re-extract	5.8	2.1

The flow ratio of solvent to feed was increased 7.9 %. The scrub to feed ratio was increased 15 %. The solvent to feed + scrub flow ratio was increased 2 %. The Tc strip flow was cut in half and the U re-extract flow was cut by almost a factor of 3. Thus, the total U product flow was cut from ~23 to ~19 mL/min.

After the last samples for test 3A were taken 8 hours after the start of hot feed, the feed point was changed initially from stage 8 to stage 10 to improve U recovery from the raffinate. Then the new flow rates were established. The change of flow rates took 3 hours to get all flows correct. Table XI shows the flow rates measured with the start time being when the feed was changed to stage 10. About an hour after the flows were adjusted a sample was taken and then another sample was taken two hours later. After the first sample at the new flow rates, the pump to recirculate chilled water to the contactors

was found to be off and the extraction stages had reached 32 °C. It is probable that higher temperatures in the extraction section could lead to higher Tc loss to the raffinate.

Table XI: Flow Rates for Test 3B with Feed in Stage 10

Run Time, hours	Solvent Flow rate, mL/min	Feed Flow Rate, mL/min	Scrub Flow Rate, mL/min	Strip Flow Rate, mL/min	U Re-Extract Flow Rate, mL/min
0.00				12.46	
0.08		4.27			
0.30		4.66			
0.30		4.58			
0.30				7.5	
0.50		4.66		8.9	
1.00				8.9	3.57
1.17				8.9	2.54
1.25			3.04	8.9	2.53
1.42			2.94	8.9	2.26
1.47				8.9	1.97
1.58	17.1				
2.50		4.53	2.91	8.90	1.75
2.58					1.83
2.67					2.07
3.00		4.53	2.13		
4.00		4.56	2.85	8.48	2.19
5.25		4.46			

The temperatures of the extraction and initial scrub stages were back to 21-22 °C by the time the second sample was taken. The remaining temperatures were given earlier in this section.

Immediately after the second set of samples was taken a large amount of solvent was observed in the raffinate decanter indicative of a hydraulic upset. The contactors were shut down, some stages drained and then restarted in an effort to resolve the problem. However, hydraulic upsets continued to occur every several hours. After about 10 hours of intermittent operation, the feed point was changed back to stage 8 and operation restarted. Table XII shows the flow rates during the final period of operation.

Table XII: Flow Rates for Test 3B with Feed in Stage 8

Run Time, hours	Solvent Flow rate, mL/min	Feed Flow Rate, mL/min	Scrub Flow Rate, mL/min	Strip Flow Rate, mL/min	U Re-Extract Flow Rate, mL/min
0.00	16.9	4.56	3.29	8.69	2.13
1.67	17.1	4.59	3.15	8.59	2.13

The change appeared to resolve the problem until another hydraulic upset occurred 4 hours after restart. Samples were taken 3 hours and 6.5 hours after returning the feed to stage 8. The test was ended shortly after the last sample when feed solution was gone. Sampling and the hydraulic upset led to fewer flow rate measurements being taken. The upsets are believed to have been the result of solids precipitating from the feed solution after earlier filtration. Solids were observed in the bottom of the feed tank toward the end of the test. Fine solids could easily be pumped to the contactor since the tank feed outlet is also at the bottom of the tank.

The results of test 3B are shown in Tables XIII through XVI.

Table XIII: Analytical Data for Test 3B with Feed in Stage 10

Stream	Run Time	U	Pu	Am	Cm	Tc	Cs	Sr
		g/L	d/m/mL	d/m/mL	d/m/mL	d/m/mL	d/m/mL	d/m/mL
U Product	3	78.0	7.58E+03	1.05E+04	2.70E+03	NA	1.62E+04	7.45E+03
	5	72.3	2.30E+03	1.68E+03	4.31E+02	NA	4.93E+03	3.35E+03
Tc Product	3	NA	1.79E+03	NA	NA	2.61E+06	1.16E+05	2.14E+04
	5	NA	1.92E+04	1.36E+04	3.49E+03	2.02E+06	3.81E+05	1.99E+05
Waste Raffinate	3	NA	NA	NA	NA	NA	NA	NA
	5	NA	1.11E+09	NA	NA	NA	NA	2.12E+10

NA - Not Available

Table XIV: Mass Balances for Test 3B with Feed in Stage 10

Stream	Run Time	U	Pu	Am	Cm	Tc	Cs	Sr
		%	%	%	%	%	%	%
U Product	3	111	3.80E-03	4.03E-03	1.21E-02	NA	3.19E-04	1.42E-04
	5	102	6.04E-04	6.42E-04	1.92E-03	NA	9.68E-05	<6.7E-05
Tc Product	3	NA	2.96E-04	NA	NA	81	1.05E-03	1.86E-04
	5	NA	3.03E-03	2.27E-03	NA	60	3.27E-03	1.65E-03
Waste Raffinate	3	NA	NA	NA	NA	NA	NA	NA
	5	NA	137	NA	NA	NA	NA	138

NA - Not Available

Table XV: Analytical Data for Test 3B with Feed in Stage 8

Stream	Run Time	U	Pu	Am	Cm	Tc	Cs	Sr
		g/L	d/m/mL	d/m/mL	d/m/mL	d/m/mL	d/m/mL	d/m/mL
U Product	3	63	2.08E+04	2.65E+02	6.79E+01	NA	NA	3.35E+03
	6.5	70	1.59E+04	NA	NA	NA	1.30E+03	3.35E+03
Tc Product	3	2.4E-04	8.4E+03	9.8E+03	2.5E+03	1.9E+06	5.5E+04	3.9E+04
	6.5	6.6E-04	1.8E+04	9.0E+03	2.3E+03	2.7E+06	1.6E+05	7.9E+04
Waste Raffinate	3	NA	NA	NA	NA	NA	NA	1.79E+10
	6.5	NA	8.24E+08	NA	NA	NA	NA	1.23E+10

NA - Not Available

The samples from test 3B (feed in stage 10) may not be at steady state due to the hydraulic upsets. Note that the U concentration in the product increased from test 3A due to lower total flow of that stream. The U mass balance is good for the last sample, but high for the first sample taken immediately prior to the hydraulic upset. The Tc concentration in the Tc product also increased in proportion to the decrease in strip flow rate. However, the mass balance is very low and appears to be decreasing. It is uncertain whether the results are due to the hydraulic upset. The data for Pu and Am appear to be high in the first sample, especially for Am. The results could reflect decreasing the number of scrub stages by two relative to test 3A; however, the second sample shows much lower Pu and Am values. The difference again could be related to the hydraulic upset that was observed immediately after the first sample. The total TRU isotopes in the U product stream for the second sample is 29 nCi per gram which makes the U lower than Class C low level waste.

Table XVI: Mass Balances for Test 3B with Feed in Stage 8

Stream	Run	U	Pu	Am	Cm	Tc	Cs	Sr
	Time	%	%	%	%	%	%	%
U Product	3	94.4	1.00E-04	1.07E-04	3.19E-04	NA	NA	6.67E-05
	6.5	97.0	5.62E-03	NA	NA	NA	2.51E-05	6.24E-05
Tc Product	3	NA	1.42E-03	1.77E-03	5.29E-03	59	5.08E-04	3.46E-04
	6.5	4.1E-04	2.83E-03	1.52E-03	4.54E-03	82	1.35E-03	6.55E-04
Waste Raffinate	3	NA	NA	NA	NA	NA	NA	135.1
	6.5	NA	118.3	NA	NA	NA	NA	93.3

NA - Not Available

The U mass balance for the portion of the test with the feed in stage 8 appears to be within expected uncertainty in flow rates and analyses. The percent of Tc in the Tc product stream is similar to the amount recovered during operation with the feed in stage 10.

Analyses for Pu in the U product stream show a large increase for the last two samples, but the Am does not increase remaining at a value similar to that observed at 4-6 hours in test 3A. The reason for change in Pu concentration is that Pu decontamination is dependent on AHA while Am decontamination is not, thus the increase is related to the AHA concentration in the scrub. These samples were taken as much as 28 hours after the last scrub was added to the scrub feed tank by mixing AHA with acid. It is known that AHA hydrolyzes in acid solutions (see equation 1).⁷



Taylor and May give a kinetic rate constant of 0.00205 L/mol-min for AHA hydrolysis at 25°C according to kinetic equation 2.

$$-d[\text{AHA}]/dt = k[\text{AHA}][\text{H}^+] \quad (2)$$

Calculation of the AHA concentration over the time of the test is shown in Figure 9. The figure shows that by the end of the test the AHA concentration was about one-third the initial concentration. The initial concentration was chosen to ensure sufficient AHA to complex Pu and reduce Np. The Pu and Np concentrations in the Dresden fuel are lower than expected for higher burnup fuels so the concentration of AHA probably didn't have to be as high as 0.47 M initially. That meant that some decomposition would not affect the Pu decontamination factor (DF). However, once the AHA concentration went below a minimum concentration, the AHA was not sufficient to maintain the Pu DF. The conclusion for the full-scale process is that AHA and HNO₃ should be fed to the contactors as separate streams rather than being mixed prior to being fed to the contactors.

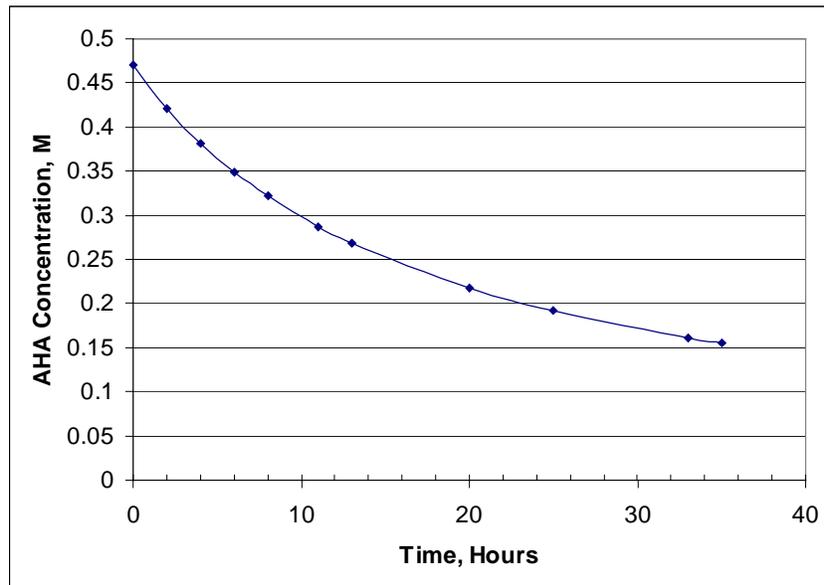


Figure 9: Calculated Decrease in AHA Concentration during Test 3

In summary, except for the TRU being high on the first test, the U meets low level waste criteria in all three tests at baseline conditions. Testing at non-baseline conditions resulted in part of the U product meeting low level waste criteria for TRU with the remainder above the limit of 100 nCi/g. Table XVII shows a compilation of results from all the tests.

Table XVII: Summary of Test Results and Low Level Waste Classes

Nuclides	LLW Limit, Ci/cubic meter			UREX Test Results ^a			
	Class A	Class B	Class C	Test 1	Test 2	Test 3A	Test 3B
Tc-99	-	-	3	0.34	0.03	NA	NA
I-129	-	-	0.08	NM	NM	NM	NM
Nuclides with half-life <5 yr	700	-	-	0.05	ND	ND	ND
Sr-90	0.04	150	7000	0.22	0.12	<0.07	<0.07
Cs-137	1	44	4600	0.47	0.41	0.04	0.03-0.11
TRU with half-life >5 yr	-	-	100 nCi/g	296	65	16	29-153
Pu-241	-	-	3500 nCi/g	1420	214	71	147-1020
Cm-242	-	-	20000 nCi/g	ND	ND	ND	ND

^a Assumes density of bulk UO₃ is half the crystal density or 3.645 g/cc

NM - Not Measured; ND - Not Detected; NA Not Available

Conclusions

Demonstration of the UREX process at baseline conditions showed that the process will meet all goals for recovery and decontamination. The goals for the UREX process are to produce a U product that is Class C low level waste or lower, recover > 99.9 % of the U and >95 % of the Tc, reject >99.9 % of the Pu to the raffinate, and the U concentration in the Tc product <0.01 % of the Tc. All three tests showed Cs being lower than the Class A limit of 1 Ci per cubic meter. Strontium was low Class B for the first two tests and very close Class A in the third test (the Sr was below the detectability limit of the method in the third test). In fact, the third test resulted in a TRU isotope concentration of only 16 nCi per gram of U. The U losses to the Tc and raffinate streams totaled 0.011 % of the U fed to the process in the first test. U loss to the raffinate was 0.016 % in the second test.

Tc losses to the U stream were 1.2 % in the first test and 0.1 % in the second test while losses to the raffinate were low in both tests. These results prove that >95 % of Tc can be recovered from the feed.

Loss of Pu and other actinides to the Tc and U product streams was <0.1 % in all first two tests of the baseline flowsheet with > 99.9 % going to the raffinate.

Future Work

Analyses of the remaining samples including stage samples from tests 2 and 3B need to be completed to give more information on the tests. The analyses need to include acid concentrations in the stage samples from the extraction and scrub section. Better analyses of the feed are needed to help clarify the mass balances Pu and Tc in the different tests. Stage samples need to be compared with calculations from the AMUSE computer code to validate the code so that the flowsheet can be optimized to obtain the best performance without extensive laboratory tests.

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